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Squeeze flow of a Carreau fluid during sphere impact

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We present results from a combined numerical and experimental investigation into the squeeze flow induced when a solid sphere impacts onto a thin, ultra-viscous film of non-Newtonian fluid. We examine both the sphere motion through the liquid as well as the fluid flow field in the region directly beneath the sphere during approach to a solid plate. In the experiments we use silicone oil as the model fluid, which is well-described by the Carreau model. We use high-speed imaging and particle tracking to achieve flow visualisation within the film itself and derive the corresponding velocity fields. We show that the radial velocity either diverges as the gap between the sphere and the wall diminishes ($Z_{tip} \rightarrow 0$) or that it reaches a maximum value and then decays rapidly to zero as the sphere comes to rest at a non-zero distance ($Z_{tip} = Z_{min}$) away from the wall. The horizontal shear rate is calculated and is responsible for significant viscosity reduction during the approach of the sphere. Our model of this flow, based on lubrication theory, is solved numerically and compared to experimental trials. We show that our model is able to correctly describe the physical features of the flow observed in the experiments. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4736742]

I. INTRODUCTION

Squeeze flows are commonplace in many engineering and biological systems and are often invoked in rheometry techniques in order to determine characteristics of both Newtonian and non-Newtonian fluids.1 In the simplest case of a parallel-plate geometry, often used for rheological measurements, where one plate is translated through a Newtonian fluid at constant velocity, exact analytical solutions of the lubrication equations can easily be derived for the radial and vertical velocity components in the film and force on the translating plate, which, neglecting slip on the solid surfaces, can be described by the Stefan-Reynolds relation

$$F = \frac{3\pi \mu D^4}{32h^3} \dot{h},$$

where $\mu$ is the dynamic viscosity of the fluid, $D$ is the plate diameter, $h$ is the gap height, and $\dot{h} = dh/dt$ is the rate-of-change of the gap height with respect to time, $t$.

Extensions of this case include modifications to the geometry, for example, the squeezing flow between a plate and a spherical lens,2,3 two rigid spheres4 or non-parallel plates.5 One can also consider the flow under a constant applied force rather than plate translation velocity or, for rheological measurements, a constant sample mass vs. constant contact area with the plates. The more practically relevant situation of the squeeze flow of a non-Newtonian fluid (e.g., power-law,6 viscoplastic, yield stress fluids,2 etc.) is considerably more complex and a thorough review of such

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flows in various geometries, fluids, slip conditions and plate translations studied previously is given by Engmann et al.\textsuperscript{1}.

Considering now the force on a sphere of radius $R_0$ in the limit as $h \to 0$, i.e., during the close approach approximation, we recover the classical lubrication force on a sphere settling toward a wall\textsuperscript{7,8}

$$F_L = 6\pi \mu R_0^2 h / h,$$

(2)

which was assessed experimentally using ultra-high-speed imaging by Marston et al.\textsuperscript{9} to provide direct verification of this force during the impact (and rebound) of solid spheres onto thin, viscous films coated on a solid wall. Mongruel et al.\textsuperscript{10} have also assessed the approach of small steel spheres toward a wall using interferometry. The impact and rebound dynamics of particles is an important consideration in wet particulate processes such as granulation, sedimentation and filtration, and has been examined both theoretically\textsuperscript{11–13} and experimentally\textsuperscript{14,15} by taking into account the elastic deformation of the particles when the sphere reaches a separation distance comparable to the lengthscale

$$x_r = \left(\frac{3\pi \theta \mu R_0^{3/2} V_0 / \sqrt{2}}{\lambda \dot{\gamma}}\right)^{2/5},$$

(3)

where $\theta$ is a parameter based on Poisson’s ratios and Young’s moduli of the two solids, $R_0$ is the sphere radius, and $V_0$ is the sphere velocity at the initial separation.

Recent developments of the original elastohydrodynamic rebound problem\textsuperscript{11} by Donahue et al.\textsuperscript{16,17} assume a constant (Newtonian) viscosity all the way down to a cut-off lengthscale, $x_{gt}$, at which point the pressure in the fluid ($O(10^5)$ Pa) induces a “glass transition.” This approach appears to neglect the shear-thinning effects in the oil films which coat the solids in the above-mentioned studies.

The specific aim of this work is two-fold; First, we extend the experimental observations of Marston et al.\textsuperscript{9} to provide direct visualisation and quantitative description of the flow field during the squeezing of a non-Newtonian (Carreau) fluid induced by the impact of a solid sphere onto a thin film. Second, a theoretical model for the flow of a Carreau fluid between a sphere and a planar surface is presented and solved numerically, which compares favourably with the experimental data.

II. EXPERIMENTAL DETAILS

A. Setup geometry

Our experimental setup is shown schematically in Figure 1(a). A steel sphere is suspended directly above the centre of the target film contained within a shallow glass tank. The sphere (Fritsch GmbH, Germany) with radius $R_0 = 20$ mm, density $\rho_s = 7800$ kg/m$^3$ and roughness $R_a = 0.06$ $\mu$m was released by an electromagnet. The tank containing the target film has inner dimensions 7 cm $\times$ 7 cm $\times$ 0.5 cm. The height of the liquid film was kept at approximately 1.5–2 mm depending on the liquid low-shear viscosity, but in some cases a thicker film up to 3.5 mm was also used. A magnified view of the impact region is shown in Figure 1(b). Here $z = 0$ indicates the base of the tank and the height of the sphere tip above the base of the tank at time $t$ is $z = Z_{tip}(t)$.

B. Fluid rheology and film preparation

The liquids used for this study were (dimethylopolysiloxane) Silicone oils (Shin-Etsu Chemical Co. Ltd, Japan) with low-shear kinematic viscosities $\nu_0 = 100$, 600, and 1000 St, with densities $\rho_l = 975$, 976, and 977 kg/m$^3$, respectively. These fluids are known to be shear-thinning and data from the manufacturer for apparent viscosity, $\mu_{app}$, versus shear rate, $\dot{\gamma}$, are shown in Figure 2. We approximate the rheological behaviour of these fluids using the Carreau model,

$$\frac{\mu_{app} - \mu_\infty}{\mu_0 - \mu_\infty} = \left[1 + (\lambda \dot{\gamma})^2\right]^{(n-1)/2},$$

(4)

where the values of $\lambda$ and $n$ are given in Table I below and $\mu_\infty = 0$. 

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In order to obtain flow-fields in the film, the fluids were seeded with 10 μm hollow glass beads (Potters Industries Inc.) with density $\rho_{hgb} \approx 600 \text{ kg/m}^3$. The large viscosity of the fluids ensures that the particle-fluid density differential did not affect the motion, which can be verified by analysing long-exposure images of a static film. The films were placed into the tanks by syringe and were left from 30 min up to several hours (depending on the viscosity) to ensure the film had reached an equilibrium level. Several films were prepared in advance for each fluid before starting experiments.

### TABLE I. Physical properties of the three silicone oils used in the experiments and parameters derived from fitting the Carreau model to viscosity vs. shear data. The values of kinematic and dynamic viscosity represent the nominal low-shear value, as stated by the manufacturers.

<table>
<thead>
<tr>
<th>Kinematic viscosity, $\nu_0$ (St)</th>
<th>Density, $\rho_l$ (kg/m$^3$)</th>
<th>Dynamic viscosity, $\mu_0$ (Pa s)</th>
<th>$\lambda$ ($\times 10^{-3}$ s)</th>
<th>$n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>975</td>
<td>9.75</td>
<td>1.15</td>
<td>0.61</td>
</tr>
<tr>
<td>600</td>
<td>976</td>
<td>58.6</td>
<td>2.91</td>
<td>0.46</td>
</tr>
<tr>
<td>1000</td>
<td>977</td>
<td>97.7</td>
<td>4.66</td>
<td>0.43</td>
</tr>
</tbody>
</table>
C. Image capture and analysis

The events were captured using a high-speed camera (Photron Fastcam SA-5) equipped with a 5× objective lens yielding a pixel resolution of 4 μm/px. Frame rates up to 30 000 fps were used, yielding time intervals Δt ≥ 33 μs, and the recording duration was sufficient to allow manual triggering. A Sumita 350 W metal halide light source with a fibre optic guide and diffuser screen was placed directly opposite the camera to achieve silhouette imaging. The video clips were saved directly to a computer, from which it was then possible to extract basic data for the gap height (between the sphere tip and the wall), Z_{tip}(t), as a function of time from impact and the inferred velocity of the sphere, V_z(t).

The temporal evolution of the velocity field in the film was analysed using PIV software (Davis 7.2, LaVision GmbH) by performing a time-series correlation between consecutive frames from the video sequences. The high-magnification used renders a narrow depth-of-field, which essentially creates a pseudo-micro-PIV technique and the field-of-view is restricted to a region extending horizontally ∼4 mm from the axis of symmetry and vertically ∼2 mm from the plate to the free-surface, i.e., z ≤ 2 mm and r ≤ 4 mm.

Multiple exposure images are presented in Figure 3 showing particle streaks over a 5-frame exposure. Here, the image has been inverted as in the first step of the PIV analysis.

Figure 4 shows the velocity field taken at time intervals of 133 μs during the close approach stage, Z_{tip} ≤ 750 μm for a steel sphere impacting onto a thin film (initial height δ = 2 mm) of the lowest viscosity oil (μ_0 = 9.75 Pa s). The sphere radius is R_0 = 20 mm and the impact velocity is V(t = 0) = V_0 = 1.5 m/s (St_0 = 2ρ_sR_0V_0/9μ_0 = 5.4). The separation distances are Z_{tip} = 728, 636, 540, 444, 356, 264, 184, and 104 μm, respectively. Here, the velocity vectors represent the absolute velocity, |v|, and the vectors themselves have been superimposed over the raw images so that the sphere edge (and hence separation distance) can be seen in each frame. No filling or interpolation has been applied to the measured fields. At a qualitative level, visual inspection of the velocity fields confirms the profiles one would expect to see under these flow conditions\(^1\) and are typical for many of the different experimental conditions realised in this work. To our knowledge, this example provides the first direct experimental velocimetry measurements of the squeeze flow of a Carreau-type fluid. We note the substantial increase in velocity magnitude as Z_{tip} decreases.
FIG. 3. Particle streaklines from multiple-exposure images during the impact of a steel sphere onto a film of the lowest viscosity oil, $\mu_0 = 9.75$ Pa s ($R_0 = 20$ mm, $\delta = 2$ mm, $V_0 = 0.63$ m/s, $St_0 = 2.24$). Original framing rate was 18,600 fps. The arrows on the left indicate the initial and end position of the sphere over the 5-frame interval. Images are taken at $Z_{tip} \approx 0.9Z_0$ and $Z_{tip} \approx 0.4Z_0$. In both, the bottom of the image represents the base of the tank.

D. Parameter space

The main dimensionless parameter assumed in previous works (and in this study) is the impact Stokes number, which gives the ratio of sphere inertia to viscous forces in the film,

$$St_0 = \frac{2\rho_s R_0 V_0}{9\mu_0}. \quad (5)$$

This parameter clearly does not incorporate the non-Newtonian effects since it assumes a single-valued viscosity, $\mu_0$. For our experiments, we use film heights, $1.5 \leq \delta \leq 3.5$ mm and impact velocities $0.63 \leq V_0 \leq 1.97$ m/s yielding Stokes numbers in the range $0.22 \leq St_0 \leq 7$ depending on the fluid low-shear viscosity. Davis et al.\textsuperscript{15} also give a critical Stokes number, above which the sphere should penetrate deep enough for deformation and rebound to occur, which is given by

$$St_c = 0.4 \ln \left( \frac{1}{\epsilon} \right) - 0.2, \quad \epsilon = 4\theta \mu_0 V_0 R_0^{3/2} / Z_0^{5/2}, \quad (6)$$

where $\theta \sim 6.15 \times 10^{-12}$ for steel onto glass and $Z_0 = 2\delta/3$ where $\delta$ is the initial film height.
FIG. 4. Velocity fields taken near the base of the tank for $z \leq 700 \, \mu m$ and $r \leq 2200 \, \mu m$ in the low-viscosity oil with $\mu_0 = 9.75 \, Pa \, s$, $\lambda = 1.15 \times 10^{-3} \, s$, and $n = 0.61$. Original framing rate was 30 kfps. Initial film height was $2 \, mm$ and the sphere radius $R_0 = 20 \, mm$. Frames shown are separated by $133 \, \mu s$. $V_0 = 1.5 \, m/s$, $S_0 = 5.4$. The colour indicates magnitude of the velocity vector.

III. THEORY

A. Model formulation

In order to model our experiments we consider the situation where a solid sphere of radius $R_0$ is approaching a solid plane as shown in Figure 1(b). The sphere is assumed to move through a thin film of oil having thickness $\delta$ and which we consider as non-Newtonian with a viscosity obeying the Carreau model given by (4). We assume that initially the sphere has penetrated the surface of the film and is at a distance $Z_0 = 2\delta/3$ from the plane and has a velocity $V_0$ pointing vertically downwards. No slip boundary conditions are applied at the surface of the sphere and plane and we assume the squeeze flow rates to be slow enough so that we may neglect inertia of the fluid and we further assume that a quasi-steady state exists at each time step as the sphere approaches the wall (see also Shuler and Advani$^{18}$ who considered a similar scenario for the flow between two plates). During the initial stages of penetration the impact Reynolds number, $Re = \rho R_0 V_0 / \mu_0$, based on
the lowest zero-shear viscosity is $O(1)$. However, in lubrication studies it is the reduced Reynolds number, $\epsilon^2 Re$, where $\epsilon = Z_{tip}/R_0$ is a small parameter, that determines whether inertial effects may be neglected (Ockendon and Ockendon\textsuperscript{19}) and this quantity is indeed small at this initial stage and becomes smaller as the sphere penetrates the film. Thus inertial effects are neglected in this work. We use a cylindrical coordinate system to formulate the governing equations, which describe the axially symmetric flow of the incompressible fluid between the sphere and the plane.

The continuity equation under such conditions may be written as

$$\frac{1}{r} \frac{\partial}{\partial r} (ru) + \frac{\partial v}{\partial z} = 0,$$

where $u$ and $v$ are the radial and axial velocities. Since we have assumed that inertial effects may be ignored the resulting momentum equation can be written as

$$\frac{\partial p}{\partial r} = \frac{\partial}{\partial z} \sigma_r,$$

where $p$ is the pressure and $\sigma_r$ denotes the shear stress. We assume that the viscosity dependence on the shear rate is captured by the Carreau model, which is described in (4). Using this expression and the assumption of squeeze flow (Equations (7) and (8)) we may write the momentum equation as

$$\frac{\partial}{\partial z} \left[ (\mu_0 - \mu_\infty) \left( 1 + \left( \frac{\lambda}{\lambda} \frac{\partial u}{\partial z} \right)^n \right) u \right] + \mu_\infty \frac{\partial u}{\partial z} = \frac{\partial p}{\partial r},$$

where the variables are as described earlier. If we allow $z = h(r, t)$ to represent the height of the sphere above the plane for a given $r$ we can apply the no-slip boundary conditions on the plane and the sphere as

$$u = 0, \quad v = 0, \quad \text{at} \quad z = 0,$$

$$u = 0, \quad v = -V, \quad \text{at} \quad z = h(r, t).$$

We may non-dimensionalise these equations using the following scales, $\bar{u} = \epsilon u/V, \bar{v} = v/V, (\bar{z}, \bar{h}, \bar{r}) = (z/Z_{tip}, h/Z_{tip}, r/R_0), \bar{\lambda} = (V/\epsilon Z_{tip})\lambda, \text{and} \bar{p} = (\epsilon^2 Z_{tip}/\mu_0 V)p$, where $V$ is the velocity of approach of the sphere, $Z_{tip}$ is the minimum height of the sphere above the plane and $\epsilon = Z_{tip}/R_0$. We note here, as explained in Sec. III B, that we are solving a system of equations under quasi-static conditions and we have that $Z_{tip}(t) = Z_0$ initially at $t = 0$, but changes as the sphere penetrates the film. Under these scalings, we have

$$\frac{\partial}{\partial \bar{z}} \left[ (1 - \bar{\mu}) \left( 1 + \left( \frac{\lambda}{\lambda} \frac{\partial \bar{u}}{\partial \bar{z}} \right)^n \bar{u} \right) \frac{\partial \bar{u}}{\partial \bar{z}} \right] = \frac{\partial \bar{p}}{\partial \bar{r}},$$

where $\bar{\mu} = \mu_\infty/\mu_0 = 0$ and

$$\frac{1}{\bar{r}} \frac{\partial}{\partial \bar{r}} (\bar{r} \bar{u}) + \frac{\partial \bar{v}}{\partial \bar{z}} = 0.$$

These equations constitute the lubrication equations for a shear-thinning fluid. The boundary conditions of the flow are

$$\bar{u} = 0, \quad \bar{v} = 0, \quad \text{at} \quad \bar{z} = 0,$$

$$\bar{u} = 0, \quad \bar{v} = -1, \quad \text{at} \quad \bar{z} = \bar{h}(\bar{r}, \bar{t}),$$

(14)

(15)
where \( h(\vec{r}, \vec{t}) = 1 + \epsilon^{-1} - \epsilon^{-1} \sqrt{(1 - \vec{r}^2)} \) is the dimensionless height of the sphere above the plane. Under the lubrication approximation we also have boundary conditions for the pressure such that

\[
\frac{\partial \tilde{p}}{\partial \vec{r}} = 0, \quad \vec{r} = 0, \quad (16)
\]

\[
\tilde{p} = 0, \quad \vec{r} = B, \quad (17)
\]

where \( B \) is the dimensionless length along the radial direction at which the pressure reaches the pressure of the surrounding and which can be viewed as the measure of the wetted portion of the sphere (see Figure 1(b)). When investigating the non-Newtonian squeeze flow between two spheres of radii \( R_1 \) and \( R_2 \), Rodin\(^{20}\) used \( B = \min(R_1, R_2) \) and Lian \textit{et al.}\(^{21}\) have shown that the most appropriate value for \( B \) is dependent on the flow index number. In the present work, we calculate \( B \) directly using the film height \( \delta \) and the minimum height \( Z_{tip} \). This means that \( B \) changes (and in general increases) as the sphere penetrates the film.

The continuity Equation (13) may be used to determine the flow rate \( \tilde{Q}(\vec{r}, t) \) along each vertical section between the plane and the sphere such that we have

\[
\tilde{Q}(\vec{r}, t) = \int_0^{h(\vec{r}, t)} \tilde{u} \cdot d\vec{z} = \frac{\vec{r}}{2}. \quad (18)
\]

B. Numerical method

We must then solve (12) and (18) together with the boundary conditions (14)–(17) to determine the velocity components and the pressure. The nonlinear nature of this set of equations means that an explicit solution for \( u \) and \( v \) is not available and we must resort to a numerical solution procedure. We use a method similar to that employed by Shuler and Advani\(^{18}\) to solve the above set of equations.

First, we discretise our domain \( 0 \leq \vec{r} \leq 1 \) into \( N + 1 \) points and similarly we divide the domain \( 0 \leq \vec{z} \leq h(\vec{r}, t) \) into \( M + 1 \) points. For a given set of parameter values for \( \mu_c, \mu_0, \lambda, \) and \( n \) we use an iterative method to find the solution of the above set of equations along each point along the \( \vec{r} \)-axis. This allows us to evaluate the pressure gradient \( \frac{\partial \tilde{p}}{\partial \vec{r}} \) and the radial velocity \( \tilde{u} \) at the \( M + 1 \) points in the \( z \)-direction for each point along the \( \vec{r} \)-direction. Integrating the pressure gradient means that we are ultimately able to determine the pressure for each of the \( N + 1 \) points at the surface of the sphere \( z = h(\vec{r}, t) \). The dimensionless force on the sphere due to the thin layer is given by

\[
\mathbf{e}_z \cdot \mathbf{F} = \tilde{F}_{\vec{z}}(\vec{z}, \vec{r}) = 2\pi \int_0^B \tilde{p}\vec{r}d\vec{r}, \quad (19)
\]

where \( \mathbf{e}_z \) is the unit vector pointing in the vertical direction. The dimensional force on the sphere may then be calculated from the relation

\[
F_{\vec{z}}(z, t) = \left( \frac{\mu_0 V_0 Z_0}{\epsilon^4} \right) \tilde{F}_{\vec{z}}(\vec{z}, \vec{r}). \quad (20)
\]

It then remains for us to solve the coupled ordinary differential equations

\[
\frac{d\tilde{v}}{dt} = \left( \frac{\mu_0 \tilde{z}_0^2}{V_0 m \epsilon^3} \right) \tilde{F}_{\vec{z}}(\vec{z}, \vec{r}), \quad (21)
\]

\[
\frac{d\vec{z}}{dt} = -\vec{v}, \quad (22)
\]

where \( m = (4/3)\pi \rho R_0^3 \) is the mass of the sphere to determine the velocity and position of the sphere as it moves through the fluid. We use a Runge-Kutta method to solve (21) and (22) with a stopping criterion based on a minimum separation distance of \( Z_{tip} = 100 \mu m \).
FIG. 5. Snapshots of the velocity field from numerical simulations ((a) and (b)) and corresponding contour plots of apparent viscosity ((c) and (d)). \( R_0 = 20 \text{ mm}, V_0 = 0.63 \text{ m/s}, \mu_0 = 58.6 \text{ Pa s}, n = 0.461, \lambda = 2.91 \times 10^{-3} \). Snapshots taken at \( Z_{tip} = 884 \mu \text{m} \) ((a) and (c)) and 112 \( \mu \text{m} \) ((b) and (d)) with \( V_z = 0.61 \) and 0.07 m/s, respectively. The vector magnitude in (a) and (b) is given by the colour bar, while arrows represent direction only.

C. Example numerical velocity field

A numerical example, showing velocity fields, is presented in Figures 5(a) and 5(b) with the corresponding (reduced) viscosity maps in Figures 5(c) and 5(d) for \( \mu_0 = 58.6 \text{ Pa s} \). The first snapshot of the vector field, at \( Z_{tip} = 884 \mu \text{m} \), exhibits a central band of high-velocity for \( r \gtrsim 3 \text{ mm} \) where \( V_r / V_z > 2 \). The vertical position of this band increases with radial position, which conforms to our expectation that \( V_{r,\text{max}} \) occurs at \( z \sim h(r, t)/2 \). We also observe that this band moves progressively toward the axis of symmetry as \( Z_{tip} \to 0 \), evident from the snapshot at \( Z_{tip} = 112 \mu \text{m} \). These flowfields clearly show us the expected flow profile, as seen in the experiments, with a high-velocity region located half-way between the sphere surface and the wall for any given radial distance.

Since the fluid here is shear-thinning, we can directly calculate the influence of the shear (induced by the local velocity gradients) on the apparent viscosity (see Sec. IV A for full details). These calculations are presented in the form of viscosity contour maps in Figures 5(c) and 5(d).
corresponding to the velocity fields shown in (a) and (b), where the apparent viscosity shows a large
distribution from \( \mu_{\text{app}} = 10 - 58.6 \text{ Pa s} \) depending on the radial and vertical location. The apparent
viscosity reduction plays an important role in determining the sphere motion and is discussed in
detail later.

IV. RESULTS AND DISCUSSION

A. Sphere motion and influence of flow index

For a direct comparison of the influence of the low-shear viscosity on the sphere motion, Figures 6(a)–6(c) present the evolution of the normalised separation gap, i.e., \( Z_{\text{tip}}/Z_0 \) vs. \( t V_0/Z_0 \), from three experiments with \( \delta \approx 2 \text{ mm} \), \( V_0 = 0.63 \text{ m/s} \), and \( \mu_0 = 9.75, 58.6 \) and 97.7 Pa s, respectively. The corresponding predicted motion from the numerical simulation is shown by the solid blue lines. In addition, to highlight the influence of the shear-thinning nature of these fluids, we have plotted the predicted decay of \( Z_{\text{tip}} \) for \( n = 1 \) (i.e., a Newtonian fluid with \( \mu = \mu_0 \)) in each plot shown by the solid red lines. The “Newtonian fluid” case in each plot clearly underestimates the penetration and a better agreement is found assuming the shear-thinning behaviour, although we observe the predicted penetration for the most viscous case, \( \mu_0 = 97.7 \text{ Pa s} \), overestimates the experimental penetration.

We note that the model assumes a quasi-steady state so that the viscosity at each time step
is adjusted based on the shear in the previous time step. Thus, the viscosity reduces essentially
instantaneously, whereas in practice, there is likely to be some delay over the short time scale of the
impact process. This may be the cause of some of the discrepancy between experiment and theory
observed in Figure 6 for the high-viscosity case.

Figure 7 further demonstrates the influence of the flow index on the maximum radial velocity,
\( V_{r, \text{max}} \), and apparent horizontal shear rate, \( \gamma = \gamma_{r_z} = V_{r, \text{max}}/h(r) \). Here, we have taken \( \mu_0 = 58.6 \text{ Pa s} \) and performed simulations for \( n = 1 \) (Newtonian) and \( n = 0.6 \) and 0.4 (shear-thinning) for comparison. The sphere comes to rest at \( Z_{\text{tip}} \approx 450 \mu \text{m} \) for the Newtonian fluid but penetrates further with \( Z_{\text{tip}} \approx 250 \mu \text{m} \) and 100 \( \mu \text{m} \) for \( n = 0.6 \) and 0.4, respectively. In Figure 7(a), we note a much more rapid drop of sphere velocity for \( n = 1 \) compared with the shear-thinning fluids (\( n = 0.6 \) and 0.4). However, more surprisingly, we find that the maximum radial velocities, \( V_{r, \text{max}} \), increase substantially during penetration for the shear-thinning fluids before decaying, whereas for the Newtonian fluid we observe only a marginal increase in \( V_{r, \text{max}} \) from 0.7–0.75 m/s between \( Z_{\text{tip}} = 1200 \) and 900 \( \mu \text{m} \), after which \( V_{r, \text{max}} \) strictly decays during the remaining penetration. In Figure 7(b) we assess the horizontal shear (see below), \( \gamma_{r_z} = V_{r, \text{max}}/h(r) \), during the approach of the sphere whereby the shear-thinning fluids show much higher shear rates with apparent rates \( \gamma_{r_z} \approx 1700 \) and 4500 s\(^{-1}\), respectively for \( n = 0.6 \) and 0.4 but only 800 s\(^{-1}\) for \( n = 1 \). The maximum apparent shear rates for the shear-thinning fluids also occur at more pronounced peaks than those obtained with the Newtonian fluid before dropping sharply during the close approach stage as \( Z_{\text{tip}} \to Z_{\text{min}} \). Note that since we use \( V_{r, \text{max}} \) in the calculation of \( \gamma_{r_z} \), the values in Figure 7(b) represent the apparent horizontal shear rate for the given radial distance.

B. Horizontal shear and apparent viscosity

For our geometry, the principal rate-of-strain is \( \gamma_{r_z} \equiv \frac{\partial V_r}{\partial z} \), which we term the “horizontal shear.” This component of shear has previously been assumed for the shear rate in the rheological equation\(^6\) for squeeze flows. Following Ardekani et al.,\(^22\) we calculate this shear rate as \( \gamma_{r_z} \approx \frac{V_{r, \text{max}}}{h(r, t)} \) where \( h(r, t) = Z_{\text{tip}}(t) + R_0 - \sqrt{R_0^2 - r^2} \) accounts for the curvature of the sphere so that \( \gamma_{r_z} = \gamma_{r_z}(r) \). A plot of \( \gamma_{r_z}(r) \) at different gap heights and different values of \( r \) is shown in Figure 8, corresponding to the realisation presented in Figure 4. Here, we can see that typical shear rates of \( \gamma_{r_z} \approx 1000 \text{ s}^{-1} \) at \( Z_{\text{tip}} \approx 720 \mu \text{m} \) increase up to \( \gamma_{r_z} \approx 10000 \text{ s}^{-1} \) at \( Z_{\text{tip}} \approx 80 \mu \text{m} \), which from the data in Figure 2, indicates a substantial reduction in the local apparent viscosity from \( \mu_0 = 9.75 \text{ Pa s} \) to \( \mu_{\text{app}} \approx 8.2 \text{ Pa s} \) at \( Z_{\text{tip}} \approx 720 \mu \text{m} \) and \( \mu_{\text{app}} \approx 4 \text{ Pa s} \) at \( Z_{\text{tip}} \approx 80 \mu \text{m} \).
FIG. 6. Normalised separation, $Z_{tip}/Z_0$, plotted against non-dimensional time, $tV_0/Z_0$, for (a) $\mu_0 = 9.75$ Pa s, (b) $\mu_0 = 58.6$ Pa s and (c) $\mu_0 = 97.7$ Pa s each with $R_0 = 20$ mm, $V_0 = 0.63$ m/s, and $\delta \approx 2$ mm. The discrete data points (+) represent the experimental data, while the solid lines represent the theoretical solutions with $n$ corresponding to the true value (blue line) and $n = 1$ (red line) for comparison.

The corresponding shear data for $\mu_0 = 58.6$ Pa s and $V_0 = 0.63$ m/s (e.g., Figure 5) are shown in Figure 9. Here we see similar initial trends as for the low viscosity ($\mu_0 = 9.75$ Pa s) example in Figure 8 whereby the shear rates increase as $Z_{tip}$ reduces from 1100 to 300 $\mu$m, but then a rapid decay as the sphere comes to rest at $Z_{tip} \approx 100$ $\mu$m. Thus for this case, we find true maximum apparent shear rates between 2000 and 3000 s$^{-1}$ depending on $r$. This translates to a local apparent viscosity between 19 and 23 Pa s, again showing a significant deviation from the low-shear viscosity value.

Further examples of the shear vs. gap height are presented in Figure 10 for a range of impact Stokes numbers (see figure caption for parameters). The data all show shear rates that span at least
two orders of magnitude, in most cases as high as $10^4 \text{s}^{-1}$ at $Z_{\text{tip}} \leq 200 \text{ pm}$, showing that localised regions of high shear occur during the close approach after impact. This shear induces reduction in the apparent viscosity, which may in turn lead to smaller gap heights than with a purely Newtonian liquid of constant viscosity. A similar conclusion regarding the shear-thinning nature and reduction of apparent viscosity was made by Marston et al.\textsuperscript{9,23} who measured the separation distance of a sphere impacting and rebounding from a solid surface covered with the same viscous oils.

Given that the apparent viscosity may be significantly reduced, a modified Stokes number based on the apparent viscosity, \( \text{St}_{\text{app}} = \frac{2\rho_s R_0 V}{9\mu_{\text{app}}} \), may be more appropriate when assessing rebound criterion for these fluids. However, since \( \mu_{\text{app}} \), given by Equation (4), is now clearly a function of the instantaneous separation height, a modified Stokes number then becomes time-dependent, evolving with the diminishing gap height. Fixing a radial position, we can then plot \( \mu_{\text{app}}(r) \) (based on the values of shear at the given radial position) as a function of the gap height. An example of this procedure is shown in Figure 11(a) for the same realisation as Figures 4 and 8. Here, we can see the local viscosity reduction is substantial for all radial positions and of a similar magnitude with apparent viscosities as low as \( \mu_{\text{app}} = 3 \text{ Pa s} \) (\( \mu_{\text{app}}/\mu_0 \sim 0.3 \)). Assuming this apparent viscosity at
FIG. 9. Shear rate, $\gamma_{rz}$ versus separation height, $Z_{tip}$, for radial positions $r = 0.5$, 1.5, and 2.5 mm. $\mu_0 = 58.6$ Pa s, $V_0 = 0.63$ m/s, $St_0 = 0.37$. Experimental data (discrete points) and theory (solid lines) are shown.

FIG. 10. Shear rate, $\gamma_{rz}$ versus separation height, $Z_{tip}$ for different impact conditions: (a) $\mu_0 = 9.75$ Pa s, $V_0 = 0.63$ m/s, $St_0 = 2.24$; (b) $\mu_0 = 9.75$ Pa s, $V_0 = 1.16$ m/s, $St_0 = 4.12$; (c) $\mu_0 = 58.6$ Pa s, $V_0 = 1.16$ m/s, $St_0 = 0.69$; (d) $\mu_0 = 97.7$ Pa s, $V_0 = 0.63$ m/s, $St_0 = 0.22$; (e) $\mu_0 = 97.7$ Pa s, $V_0 = 1.16$ m/s, $St_0 = 0.41$; (f) $\mu_0 = 97.7$ Pa s, $V_0 = 1.55$ m/s, $St_0 = 0.55$. See legends for radial positions.
FIG. 11. (a) Apparent viscosity, $\mu_{\text{app}}$, plotted as a function of sphere separation for $r = 0.5$ and 2 mm. The values of $\mu_{\text{app}}$ are calculated from the maximum apparent shear rates shown in Figure 8 and Equation (4), $\mu_0 = 9.75$ Pa s, $V_0 = 1.5$ m/s, $St_0 = 5.4$. (b) Apparent Stokes number (based on apparent viscosity $\mu_{\text{app}}$) versus normalised separation height based on apparent viscosities in (a).

Each time step (or gap height), we can then plot a modified Stokes number based on this apparent viscosity, shown in Figure 11(b) for $r = 2$ mm. Here we can clearly see that the instantaneous ratio of sphere inertia to viscous forces actually increases during the penetration, despite the decay of $V_z$.

If we seek the maximum values of this modified Stokes number, based on the maximum viscosity reduction, we can assess the peak ratio of sphere inertia to viscous forces during the approach stage. This procedure, in general, results in an increase in the Stokes number from the impact value, $St_0$, as shown in Figure 12, where we can see that $St_{\text{app}} > St_0$ for most cases. This clearly highlights the dramatic reduction in apparent viscosity. We note, however, that the values of $\mu_{\text{app}}$ are calculated from maximal values of $\gamma_{rz}$, rather than some average value (since the shear is a function of both radial position and sphere penetration depth), so that this procedure estimates the maximum value of $St_{\text{app}}$. 
C. Elasticity lengthscale

The vertical separation at which the lubrication force becomes significant enough to cause solids deformation, i.e., the elasticity lengthscale, is given by Davis et al.\textsuperscript{15} as \( x_r = (3\pi \theta \mu R_0^3 / 2 V_0 / \sqrt{2} )^{2/5} \). It is only when the separation between the sphere tip and wall reaches this lengthscale that rebound is observed. However, this lengthscale does not account for the shear-thinning effect observed with silicone oils. Since we now have experimental data for \( V_z \) and estimates for \( \mu_{\text{app}} \), we can use these values to provide a refined elasticity lengthscale, \( x_{\text{min}} = (6\pi \theta \mu_{\text{app}} R_0^3 / 2 V_z / \sqrt{2} )^{2/5} \), which

\[ \mu_0 = 9.75 \text{ Pa.s} \]
\[ \mu_0 = 58.6 \text{ Pa.s} \]
\[ \mu_0 = 97.7 \text{ Pa.s} \]

FIG. 12. Modified Stokes number, \( St_{\text{app}} \), plotted against the impact Stokes number, \( St_0 \). Error bars encompass multiple trials. The dashed line indicates a slope of 1, i.e., where \( St_0 = St_{\text{app}} \).

FIG. 13. Predicted minimum separation based on apparent viscosities from Figure 11. The value of \( x_{\text{min}} \) has been normalised by \( x_r = 19.6 \mu \text{m} \).
corresponds to the expected minimum separation based on \( \mu_{app} \) and \( V_z \). Using the instantaneous values for both \( V_z \) and \( \mu_{app} \), we can then plot the “updated” value for \( x_{min} \). In Figure 13 we plot data for \( x_{min} \) for the same data shown in Figure 11, showing that \( x_{min} \approx 19.6 \mu m \) for \( Z_{tip} \approx Z_0/3 \) but that \( x_{min} \sim x_r/2 \approx 9 \mu m \) during close approach when \( Z_{tip}/Z_0 \leq 0.1 \). With this approach, and apparent viscosities used in Figure 12, we find reduced lengthscales for all experimental trials shown in Figure 14 with many reduced by about a half. This should certainly be taken into account when assessing rebound criteria for particles in non-Newtonian fluids.

V. CONCLUSIONS

In summary, we have conducted experiments to examine the flow-field in the region directly beneath a sphere, which impacts onto a thin, viscous film of oil, creating a squeeze flow. Using high-magnification and narrow depth-of-field, we have performed pseudo-micro-PIV at frame rates up to 30 kfps, which provides direct measurements of the velocity field and enabled calculation of the principal deformation rate—the horizontal shear.

The experimental results clearly show that there are high-shear regions \( (\gamma_r (r) \approx 10^4 \text{ s}^{-1}) \) close to the axis of symmetry with \( r/R_0 \approx 0.1 \), which, owing to the non-Newtonian nature of the oils, inevitably leads to significant viscosity reduction, up to 60% in some cases. This viscosity reduction may in turn lead to increased sphere penetration, as indicated by the “updated” elasticity lengthscale, \( x_{min} = (6\pi \theta \mu_{app} R_0^3 V_z / \sqrt{2})^{1/3} \), incorporating the instantaneous sphere velocity and apparent viscosity, which can reduce by up to a half for some realisations. As such, it appears the impact Stokes number, taking a single-valued viscosity, is the correct non-dimensional number to characterise the impact and penetration for Newtonian fluids, but for shear-thinning fluids, a reduced viscosity value may be taken to estimate the deeper penetration observed.

This increased penetration may be responsible for the rebound-induced cavitation at sub-critical Stokes numbers (i.e., for cases where \( S_{tip} < S_t \)). This finding needs to be taken into account in mathematical models of the impact-rebound process, which is an important consideration for simulating agglomeration rates in granulation.
Our model of this physical situation, which incorporates the non-Newtonian effects by employing a Carreau model for the apparent viscosity, was solved numerically and compared favourably to the experimental results. While some aspects of the model may be improved, for example, by taking into account a model to determine the deformation of the free surface as the sphere penetrates the film, in order to improve the quantitative agreement, overall the model is able to replicate many of the features of the flow observed in the physical experiments and may be useful in more complex models of higher order processes in multi-particle systems. Extensions of this model to assess the possible compressibility effects highlighted by Marston et al. are the subject of ongoing work.

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